Electrochemistry Communications 45 (2014) 67–70

Contents lists available at ScienceDirect

Electrochemistry Communications

journal homepage: www.elsevier.com/locate/elecom

Short communication

A new upgraded biogas production process: Coupling microbial electrolysis cell and anaerobic digestion in single-chamber, barrel-shape stainless steel reactor

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ARTICLE INFO

Article history:
Received 30 April 2014
Received in revised form 16 May 2014
Accepted 20 May 2014
Available online 28 May 2014

Keywords:
Upgraded biogas
Carbon dioxide
MEC–AD coupling system
Stainless steel reactor

ABSTRACT

Production of upgraded biogas is required to remove as much carbon dioxide as possible. It was found that by coupling microbial electrolysis cell (MEC) and anaerobic digestion (AD) in a single-chamber, barrel-shape stainless steel reactor, compared with common anaerobic digestion (control), CH_4 content in excess of 98% was achieved and CH_4 yield was increased 2.3 times. Meanwhile, the COD removal rate was tripled and carbon recovery was increased by 56.2%. In this new process, unwanted CO_2 was *in situ* converted into CH_4 on anode by the dominant microbes, hydrogenotrophic electromethanogens (*e.g. Methanospirillum*). These microbes could utilize hydrogen gas generated at the inner surface of stainless steel reactor, which itself served as cathode of MEC through small voltage addition (1.0 V). The overall energy efficiency was 66.7%.

1. Introduction

Biogas derived from anaerobic digestion (AD) of organic waste is primarily a mixture of methane (CH_4, 50–75%) and carbon dioxide (CO_2, 25–50%). The main chemical equation for the biogas production is: $CH_3COOH \rightarrow CO_2 + CH_4$. It is well known that the unwanted CO_2 will reduce the quality of biogas and contribute to negative effect on biogas compression. Biogas, once purified by removing the impurities, mainly CO_2, can be used as renewable and low carbon fuel substituting natural gas for electricity generation and natural gas vehicle transportation. Methods for biogas upgrading is mainly focused on the removal of CO_2 accompanied with a little CH_4 loss [1]. Nevertheless, the strategy for upgraded biogas production by *in situ* converting CO_2 into additional CH_4 and increasing CH_4 yield simultaneously has never been reported.

Rapidly developing bioelectrochemical technology has been proved to be a promising platform for CO_2 capture and conversion comparing with other methods [2]. With a small addition of voltage at the microbial electrolysis cell (MEC), electromethanogens can use electrons or hydrogen formed at cathode to convert CO_2 into CH_4 directly [3,4]. In the present work, a novel process, *i.e.*, coupling microbial electrolysis cell and anaerobic digestion, for *in situ* converting CO_2 into CH_4 and enhancing CH_4 production simultaneously was introduced. In this new process, the chemical equation for the biogas production can be simplified as follows: $CH_3COOH \rightarrow 2CH_4$, which has never been documented.

Stainless steel has been considered to be cost–benefit catalyst for hydrogen evolution and material for scaling-up [5,6], thus the reactor was constructed by stainless steel which also served as the cathode of MEC in this study. Voltages were applied to study the variations of CO_2 capture and conversion efficiency and CH_4 yield first. The mechanisms for upgraded biogas production in stainless steel reactor were then investigated.

2. Materials and methods

2.1. Reactor setup

Single-chamber, barrel-shape reactor with total volume of 180 mL (diameter: 5.0 cm; height: 9.2 cm) was made of stainless steel (SUS304), which itself was used as cathode. Anode was carbon felt of 2.0 × 5.0 cm pretreated as previously described in ref. [7]. Titanium wires were used to connect the electrodes to the circuit. Electrode of Ag/AgCl (sat. KCl, 0.197 V *vs.* standard hydrogen electrode, SHE) was used as reference electrode.

2.2. Experiments and measurements

The experiments were conducted in three reactors (AD, E1 and E2). The reactor AD was an anaerobic digested reactor, circuit open, without any energy input. Reactors E1 and E2 were inoculated methanogens

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http://dx.doi.org/10.1016/j.elecom.2014.05.026
1388-2481/

derived from the solution of an existing two-chamber MEC reactor [8] that was producing methane. A power source (DC Power Supply GPD-4303S, Taiwan) was connected to the circuit of E1 and E2 to add fixed voltage of $E_{ap} = 0.2$ V, and a multimeter (2700; Keithley Instruments, Inc.) was used to monitor the voltage across an external resistor ($R_{ex} = 10\ \Omega$) for current calculating. The two reactors were operated for three months with acetate (1 g/L) in a buffered nutrient medium which was detailed in ref. [9]. After the acclimation, batch tests were conducted as follows: 20 mL waste activated sludge (pH 6.8, total suspended solids (TSS) 1.75×10^4 mg/L, volatile suspended solids (VSS) 1.38×10^4 mg/L) was inoculated into each reactor (AD, E1 and E2) containing 150 mL same nutrient medium using acetate (10 g/L), the typical end-product of carbohydrate or protein fermentation, as carbon source. Reactors E1 and E2 were then operated at the added voltages of $E_{ap} = 0.4$ and 1.0 V, respectively. In addition, two abiotic anode MEC–AD coupled reactors AR1 and AR2 were set up applying voltages of 0.4 and 1.0 V, respectively. 2-Bromoethanesulfonate (50 mmol/L) was applied to both reactors to inhibit methanogen metabolisms. pH value in these 5 reactors was adjusted to pH 7.0 ± 0.1. All reactors were sealed with rubber stoppers and gas was collected in a 0.5 L gas bag. All experiments were conducted in triplicate at a constant-temperature (30 °C) without pH control.

2.3. Analysis and calculation

Gas (CH_4, CO_2 and H_2) and acetic acid (CH_3COOH) were detected in accordance with ref. [8]. Microbial samples scraped from the same anode biofilm at three different sites were mixed together for DNA extraction. Then MiSeq sequencing of 16S rRNA gene amplifications were conducted to characterize the membership and function of microbiome [10,11]. Cyclic voltammetry (CV) was conducted in the potential range from −0.8 to 0.4 V at a low scan rate of 1 mV/s.

Carbon recovery based on total mole carbon of CH_4 recovered compared to the initial mole carbon of substrate. Overall energy efficiency relative to both electrical input and energy of the substrate was evaluated as previously described in ref. [12].

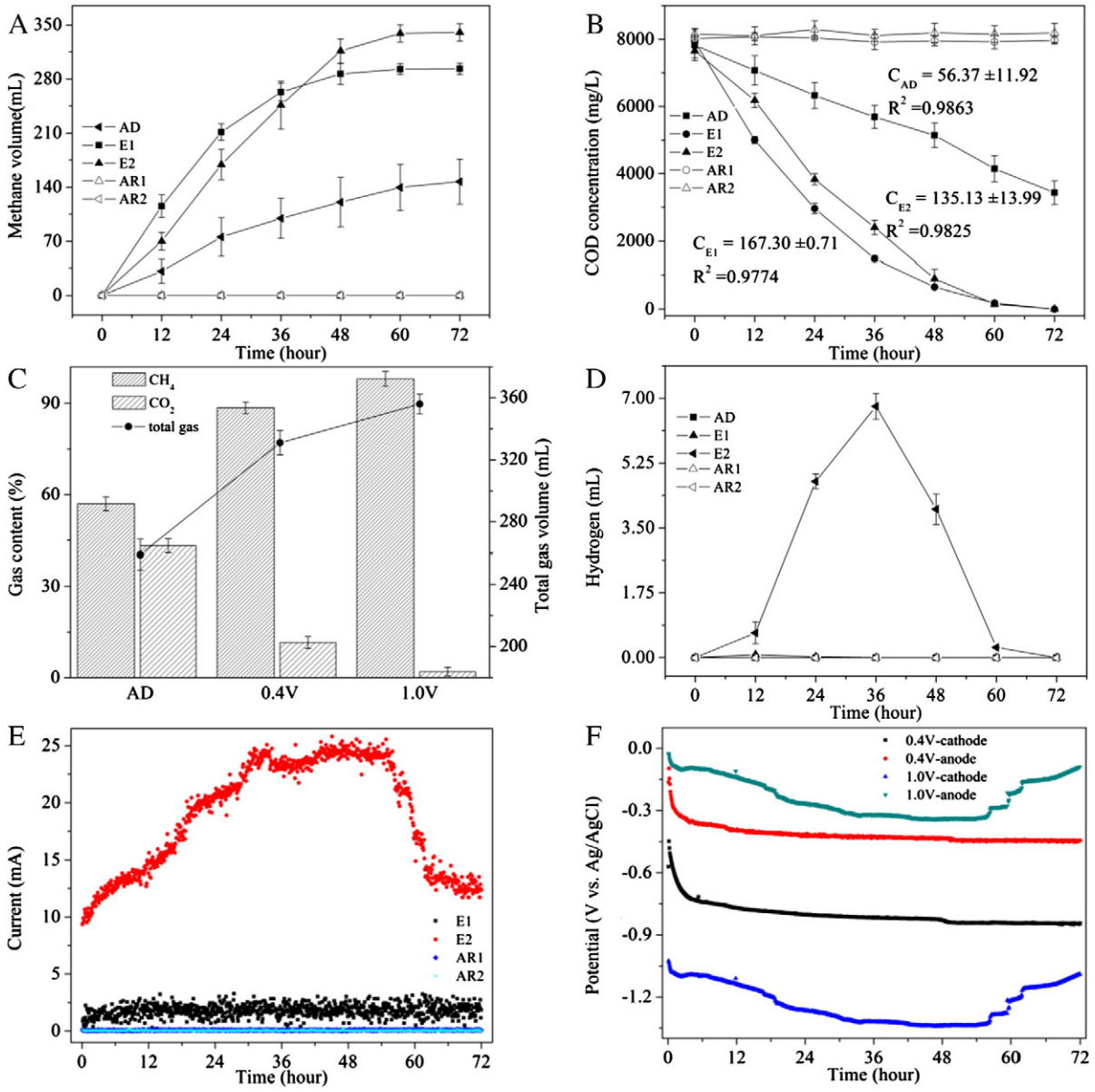

Fig. 1. (A) Comparison of methane yields at different reactors. (B) Comparison of COD removal efficiency at different reactors. (C) Changes of CO_2 and CH_4 contents at different reactors. (D) Hydrogen generation at different reactors. (E) Current generation at different reactors. (F) Changes of cathode and anode potential with time at different applied voltages (AD, anaerobic digestion reactor; E1, applied voltage of $E_{ap} = 0.4$ V; E2, applied voltage of $E_{ap} = 1.0$ V; AR1, control with applied voltage of $E_{ap} = 0.4$ V; AR2, control with applied voltage of $E_{ap} = 1.0$ V).